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LABORATORY STUDIES ON THE ADSORPTION OF RADIOIODINE AND IODINE COMPOUNDS ON ACTIVATED CARBON

R. Schwarzbach

Foreign Technology Division
Wright-Patterson Air Force Base, Ohio

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LABORATORY STUDIES ON THE ADSORPTION OF RADIOIODINE AND IODINE COMPOUNDS ON ACTIVATED CARBON

R. Schwarzbach*

Activated carbons produced in the German Democratic Republic are tested with regard to their capability to adsorb radioiodine and radioactive methyl iodide. The physico-chemical properties of the available types of activated carbon are summarized. Activated carbons with most favorable properties are selected, considering different parameters. It is attempted to improve the adsorptive properties of the selected carbons by preparing them with $Cu(NO_3)_2$, $AgNO_3$ or KI. Furthermore, the influence of water vapor, temperature and carbon dioxide on the effectiveness of the carbon was tested. Methods and results of the tests are presented.

1. Statement of the Problem

With the establishment of many nuclear technology stations, the importance of protecting the environment from the output of radioactive materials into the atmosphere is growing. The problematic of the behavior of radioiodine, its radiotoxicity, the nature of its elimination, especially during damage situations, make it the object of many research programs and studies. [1-5]

Iodine appears in several physical-chemical forms in the atmosphere of nuclear plants: 1. bound to aerosols; 2. in the elemental form as iodine vapor; and 3. also in organic compounds (mostly as methyl iodide). As there are suitable methods and equipment for aerosol separation, here we investigate only the

* Central Institute for Nuclear Research Rossendorf Works performed for the Federal Central Bureau for Radiation Protection, East Germany.

behavior of radioiodine in the form of iodine vapor and of organic iodine in the form of methyl iodide.

Various methods may be used for iodine removal. High decontamination factors can be attained with physical processes, such as freezing out, adsorption on solids such as silica gel, molecular sieves, Al_2O_3, absorption in washing solutions or on materials impregnated with thiosulfate, silver nitrate, potassium iodide, tertiary amines, etc. The adsorbent most often used today in nuclear plants for separating radioiodine from the exhaust gas is activated carbon [6, 7, 8]. The activated carbons provided for iodine testing were products of the German Democratic Republic (Peoples Synthesis Plant, Schwarzheide: WDG 010, WDK 14, WDK 14F; and the "Friedrich Engels" synthetic fiber plant at Premnitz: R3, R4 and AS). The activated carbons were evaluated by measurements of the filtration effectiveness, the filtering capacity, the strength of the carbon, the flow resistance, the flammability, preparation with various chemicals to improve the adsorption properties, the desorption behavior, etc.

2. Experimental

The apparatus sketched (Figure 1) was used for testing activated carbon with elemental iodine as well as with methyl iodide. The flow of carrier gas (air) moved from the steel cylinder through a rotameter into the vessel in which iodine or methyl iodide was developed. Iodine was made by reaction of $Na_2Cr_2O_7$ with carrier-free $Na^{131}I$, and passed into the attached cooling vessel. The methyl iodide was generated according to the equation:

$$(CH_3)_2SO_4 + 2\ Na^{131}I \rightarrow 2\ CH_3{}^{131}I + Na_2SO_4$$

and was likewise deposited in the cooling vessel. By thermostatic control of the cooling vessel, an amount of $^{131}I_2$ or $CH_3{}^{131}I$

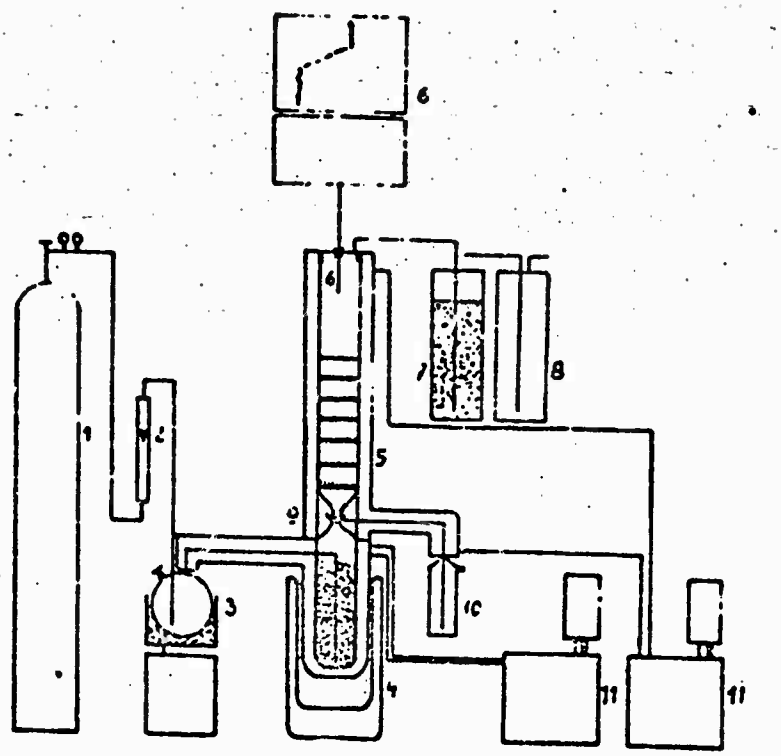

Figure 1. Apparatus for determining the adsorption of $^{131}I_2$ or $CH_3{}^{131}I$ on activated carbon.

1. Steel cylinder. 2. Rotameter. 3. Iodine generation vessel and control transformer. 4. Cooling vessel. 5. Filter being measured. 6. Gas flow counter tube with measuring point and recorder. 7. Safety filter. 8. Wash bottle. 9. Three-way cock. 10. Sampling bottle. 11. Thermostat.

corresponding to the temperature was vaporized out of the cooling vessel and led by the carrier gas flow through the activated carbon test bed. In order to check the activity during the experiment, a gas flow counting tube is connected following the filter, with a pulse rate meter and a recorder. The following counting tube proved to be good, because any filter leaks could be noted immediately because of the rapid rise in activity. After the activated carbon bed there was a safety filter, consisting of a washing bottle with thiosulfate solution, for complete removal of radioiodine or methyl iodide. The cooling vessel and the activated carbon filter could be heated separately with 2 thermostats. A three-way cock ahead of the activated carbon filter allowed the gas flow to be taken off for separate checking. No iodine activity could be demonstrated in the thiosulfate washing bottle after all the experiments. The actual evaluation of the activated carbon samples was done with a NaI scintillation crystal at 0.36 MeV, outside the apparatus shown.

3. Experimental Results and Discussion

3.1 Physical-chemical Parameters of the Activated Carbon

The surface areas of the activated carbons were determined with N_2 by the BET method. The results are shown in Table 1. Table 2 shows the water content of the commercial product, which was determined by weight loss after drying at 110°C. The AS carbon had the lowest water content. The residue after ignition, determined according to TGL 9493, is also shown in Table 2.

The Type WDG 010 activated carbon had more than a fourth of its weight as uncombustible residue. Determination of the carbon dusting showed that the WDG 010 activated carbon had low strength, and 50% carbon dust was found in WDG 010 when the particle size distribution was checked by sieve analysis, while the R3, R4, and AS contained less than 0.25% carbon dust. If one considers

TABLE 1. DETERMINATION OF THE SURFACE AREA BY THE BET METHOD

Carbon Type	Surface (m^2/g)
AS	1265
B3	1130
B4	710
WDK 14 P	810
WDK 14	765
WDG 010	620

TABLE 2. DETERMINATION OF THE WATER CONTENT AND RESIDUE ON IGNITION

Carbon Type	Water Content (%)	Residue on ignition (mg/g carbon)
S	4,8	81,0
3	15,8	40,6
4	12,3	76,4
DK 14 P	50,7	18,0
DK 14	42,7	41,7
DG 010	49,8	264,7

the results of these studies, it appears that the AS activated carbon has a desirable behavior with respect to the dusting resistance, water content, and residue on ignition, and has the greatest surface.

2. Measurements of the Adsorption Behavior of the Activated Carbon With Respect to Macro-Amounts of Iodine or Methyl Iodide

In order to determine the adsorption capacity of the activated carbons under static conditions, 1 g portions of activated carbon and excess iodine or methyl iodide were placed in a sealable vessel at a constant temperature of 80°C or 23°C, respectively. The course of adsorption was followed gravimetrically to constant weight. Table 3 shows the amounts of iodine or methyl iodide adsorbed, expressed in grams of iodine or methyl iodide per gram of carbon. Vogt and Meringdal [6] report similar values of 2 to 4 grams of iodine per gram of carbon.

The adsorption capacity under dynamic conditions was measured at 80°C in the apparatus sketched in Figure 1, with 1g of carbon using an incident flow velocity of 2.1m/minute for iodine. Methyl iodine adsorption was determined by means of the retention time, using gas chromatography (see Table 4). Due to the high adsorption capacity of the activated carbon, inactive iodine was used for testing. The amounts of iodine adsorbed on the activated carbons are shown in Table 5. Under the test conditions selected, the amounts adsorbed are about 1 g iodine per gram of carbon. These values are not to be considered in practice, though, because a filter cannot be used to zero efficiency, especially in nuclear plants. The actually usable filter capacity could be considerably below 0.1 g iodine per gram of carbon. From the view point of the nearly carrier-free occurrence of radioiodine in nuclear plants, this gives enormous useful lifes. These filter lifes are reduced by the presence of still other adsorbable foreign gases or even by the inactive iodine content of 10^{-7} g/m^3 [9] in the atmosphere. With respect to activity, the filter can be loaded to about 1 Ci/g carbon. Desorption phenomena are still negligible at this activity concentration [7].

TABLE 3. STATIC ADSORPTION OF IODINE ON ACTIVATED CARBON: AT 80°C, 1 g DRIED CARBON, DRY ATMOSPHERE

Carbon Type	Amount of Iodine Adsorbed (g iodine/g carbon)
AS	2,06
R3	1,82
RA	1,46
WIK 14 P	1,22
WIK 14	1,16
WDG 010	0,88
AS + $AgNO_3$ Impregnation	1,98
R3 + $AgNO_3$ "	1,74
RA + $AgNO_3$ "	1,42
WIK 14 P + $AgNO_3$ "	1,20
WIK 14 + $AgNO_3$ "	1,12
WDG 010 + $AgNO_3$ "	1,06

STATIC ADSORPTION OF METHYL IODIDE ON ACTIVE CARBON: AT 23°C, 1 g DRIED CARBON, DRY ATMOSPHERE

Carbon Type	Amount of Iodine Adsorbed (g iodine/g carbon)
WDG 010	1,72
WIK 14	1,69
WIK 14 P	1,51
AS	1,26
R3	1,09
RA	0,88
AS + $Cu(NO_3)_2$ Impregnation	1,28
AS + $AgNO_3$ "	1,24
AS + KI "	0,86

TABLE 4. DYNAMIC ADSORPTION OF METHYL IODIDE ON ACTIVATED CARBON: 1 g CARBON, 80°C, INCIDENT FLOW RATE 1.06 m/min.

Retention times in minutes, determined by gas chromatography							
Carbon Type	Untreated	CO_2	$AgNO_3$	$Cu(NO_3)_2$	Heat 470°C	Water Vapor	KI
WIK 14 F	39,1	35,9	42,1	40,4	24,1		2,8
WIK 14	34	30,4	36,2	34,4	32,2		2,2
AS	32	35	32,3	27,6	16,4	42,8	6
R3	24	14,9	20,2	16,5	11,8	17,4	3
WDG 010	13	10	11	11	17,1	12,4	1,6
RA	11	6,8	7,1	8,4	7,5	8,8	2,3

TABLE 5. DYNAMIC ADSORPTION OF IODINE ON ACTIVATED CARBON AT 80°C, INCIDENT FLOW RATE 2.1 m/min, 1 g CARBON.

Amount of iodine adsorbed in g iodine/g carbon				
Carbon Type	Untreated	Impregnated with $AgNO_3$	Impregnated with $Cu(NO_3)_2$	Tempered at 470°C
AS	1,98	0,26	1,12	2,13
R3	1,66	0,35	0,97	1,57
RA	1,09	0,22	1,06	1,21
WIK 14 F	0,93	0,21	0,53	1,24
WIK 14	0,90	0,29	0,52	0,97
WDG 010	0,87	0,21	0,46	0,89

3.3 Measurements of the Adsorption Behavior of Activated Carbons With Respect to Micro Amounts of Iodine and Methyl Iodide

Under the conditions selected, the tests with carrier-free ^{131}I iodine and carrier-free methyl iodide gave high degrees of separation for both adsorbates. The filter efficiency was

determined from the ratio of the activity concentration on the measured filter to the total activity concentration:

$$\eta = \frac{A_1}{A_1 + A_2}$$

A_1 = activity concentration of the measured filter

A_2 = activity concentration of the residual filter

The residual filter was dimensioned so that there could be complete separation of the radioiodine or methyl iodide. Testing of the following wash bottle showed no activity. Contamination losses could be kept very small by using a glass apparatus and test temperatures of 80°C. This favorable degree of separation, even for methyl iodide, is characteristic of a dry atmosphere, while lower separation for methyl iodide is to be expected for air saturated with water vapor [10].

From Table 6 we can see that the activated carbons WDG 010 and AS had good behavior for carrier-free iodine and methyl iodide. Taking the physical properties of the activated carbons into consideration, the AS carbon was used for further experiments with the goal of improving adsorption by impregnating or activating. The retention times determined by gas chromatography are shown in Table 4. Some of the different preparations gave improvements in the retention times, especially for the $AgNO_3$ impregnation. The activated carbons from the German Democratic Republic production showed adsorption capacities and degrees of separation which were very similar to the Type H 32 activated carbon from the German Federal Republic, which is being used for removal of radioiodine from the waste gas streams of nuclear plants. As the tests for characterizing the adsorption behavior of the activated carbons from the German Democratic Republic production are being continued, no final evaluation can yet be given.

TABLE 6. ^{131}I ADSORPTION, CARRIER-FREE AT 80°C, 1 g CARBON, INCIDENT FLOW VELOCITY 2.1 m/min

Carbon Type	%
AS	99,47
R3	79,93
RA	99,58
WDK 14 P	95,5
WDK 14	98,57
AS + KI	99,33
AS + $AgNO_3$	99,65
Ag + $Cu(NO_3)_2$	99,77

$CH_3{}^{131}I$ ADSORPTION, CARRIER-FREE AT 80°C, 10 g CARBON, INCIDENT FLOW VELOCITY 0.24 m/min

Carbon Type	%
AS	99,66
R3	99,98
RA	99,08
WDK 14 P	99,96
WDK 14	97,81
AS + KI	99,84
AS + $AgNO_3$	99,93
Ag + $Cu(NO_3)_2$	99,68

Although most nuclear plants use activated carbons because of their high adsorption capacity, their flammability at high temperature cannot be overlooked. Various authors were able to increase the ignition temperature to 500°C with special types [11]. Along with improvement of the adsorption properties of activated carbon by impregnation and activation, there are recent literature reports on the search for other adsorber materials suitable for separating radioiodine from nuclear plants [12, 13, 14].

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